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ON IMPROVED 3-D NON-GAUSSIAN NETWORK MODELS FOR RUBBER ELASTICITY

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Introduction

Network theories of rubber elasticity are generally considered to be well-established by now. The non-Gaussian statistical mechanics model for the large strain extension of a single long-chain molecule dates back to the 1940's; modeling the response of a network of n randomly oriented chains (per unit volume) in terms of three independent sets of $n/3$ such chains in three orthogonal directions was suggested by Wang and Guth [1]. This simple approach has become quite popular since, although it is rather approximate as demonstrated later by Treloar for uniaxial tension and biaxial tension with fixed principal stretch directions (see e.g. [2], [3]). This simple 3-chain modeling has very recently gained interest also in the constitutive modeling of large inelastic deformations of glassy amorphous polymers. As suggested for instance by Haward and Thackray [4], the orientation hardening observed during straining of an amorphous glassy material after plastic flow has been initiated, is primarily a result of the straightening out of polymer chains between physical entanglements. Accordingly, this type of hardening has been modeled by Boyce, Parks and Argon [5] by effectively incorporating the above-mentioned 3-chain non-Gaussian network model into their constitutive theory. However, very recently, Arruda and Boyce [6] found that this model was not capable of picking up the strain hardening observed experimentally in polycarbonate. At the same time, they suggested to model the network by eight equivalent chains instead of three, and obtained much closer agreement with their experiments.

Comparing these 3- and 8-chain representations with the actual 3-D initially random distribution of molecular chains, we intuitively felt that the 3-chain model would be likely to overestimate the actual stiffness of the network, while the 8-chain representation would probably give a lower bound. Therefore, we study here a more accurate non-Gaussian rubber model in which full account is taken of the orientation distribution of the individual chains in the network. The present treatment closely follows that in [3], but extends the latter to general 3-D deformations including arbitrary rotations of the principal stretch axes. The accuracy of the 3- and 8-chain models is assessed

for a number of deformation processes by solving the governing equations numerically to a high degree of precision. Finally, a simple interpolation is proposed which captures the numerical results very accurately.

Full Network Model

Following the classical work of Treloar [2], the present theory assumes the network to contain n randomly jointed chains per unit volume, each containing N links of length l . It is further assumed that, in the unstrained state of the network, the end-to-end vector \mathbf{r}_0 for each chain has the vector length r_0 corresponding to the root-mean-square length $\sqrt{N}l$ for the free chain, and that the vectors \mathbf{r}_0 are randomly oriented in space. When this network is subjected to some three-dimensional deformation process represented by the deformation gradient tensor \mathbf{F} , each end-to-end vector \mathbf{r}_0 is taken to be distorted (i.e. strained) and rotated to the vector \mathbf{r} in an affine manner following the usual way. The associated principal stretches λ_i ($i = 1, \dots, 3$) as well as the corresponding Lagrangian principal directions \mathbf{e}_i^0 in the undeformed configuration and the Eulerian principal directions \mathbf{e}_i in the current deformed configuration are defined through $(\mathbf{F}^T \mathbf{F}) \mathbf{e}_i^0 = \lambda_i^2 \mathbf{e}_i^0$ and $(\mathbf{F} \mathbf{F}^T) \mathbf{e}_i = \lambda_i^2 \mathbf{e}_i$, respectively. Due to the affine deformation assumption and the isotropy of the initial orientation distribution of the chain vectors \mathbf{r}_0 , the total free energy W for the network can be expressed solely as a function of the principal stretches λ_i . By a standard argument (see, e.g. [7]), it then follows that the principal axes of the Cauchy stress tensor $\boldsymbol{\sigma}$ coincide with the Eulerian triad, so that

$$\boldsymbol{\sigma} = \sum_{i=1}^3 \sigma_i (\mathbf{e}_i \otimes \mathbf{e}_i) \quad (1)$$

with the principal stresses σ_i required to sustain the imposed deformation being given by

$$\sigma_i = \lambda_i \frac{\partial W}{\partial \lambda_i} - p \quad (\text{no sum}). \quad (2)$$

Here, p is an additional hydrostatic pressure which is left indeterminate by the free energy function because of incompressibility, $\lambda_1 \lambda_2 \lambda_3 = 1$, but which is determined from the boundary conditions.

We start out by briefly recapitulating the well-known considerations for a single chain [2], [3]. Consider a single chain with its end-to-end vector \mathbf{r}_0 in the unstrained state specified by angular coordinates θ and φ with respect to the Lagrangian triad as shown in Fig. 1. The components of the unit direction vector $\mathbf{m} = \mathbf{r}_0/r_0$ on the Lagrangian axes \mathbf{e}_i^0 then are

$$m_1 = \sin \theta \cos \varphi, \quad m_2 = \sin \theta \sin \varphi, \quad m_3 = \cos \theta.$$

Invoking the affine deformation assumption, the stretch $\lambda = r/r_0$ of the corresponding end-to-end vector in the strained state can be given by

$$\lambda^2 = \sum_{i=1}^3 \lambda_i^2 m_i^2.$$

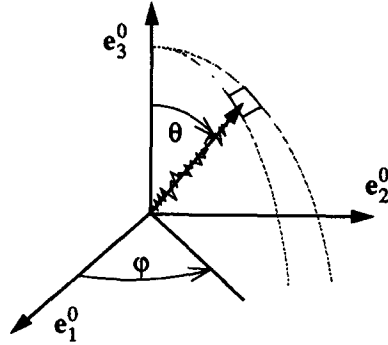


FIG. 1

A single chain in the unstrained state; definition of geometric quantities

According to the non-Gaussian statistical mechanics network model of Wang and Guth [1], the Helmholtz free energy w for a single chain at stretch λ (at constant absolute temperature T) is determined by the configurational entropy s of the chain, $w = -Ts$, which is given by

$$w = kNT \left[\frac{\lambda}{\sqrt{N}} \beta + \ln \frac{\beta}{\sinh \beta} \right] - w_0, \quad \beta = \mathcal{L}^{-1} \left(\frac{\lambda}{\sqrt{N}} \right), \quad (3)$$

where w_0 is a constant, k is Boltzmann's constant and \mathcal{L} is the Langevin function defined by $\mathcal{L}(\beta) = \coth \beta - 1/\beta$.

Now consider a network with n chains (per unit volume) with a random distribution of the orientation of their end-to-end vector \mathbf{r}_0 in the unstrained state. The number of chains whose \mathbf{r}_0 -vector orientation falls in the range between (θ, φ) and $(\theta + d\theta, \varphi + d\varphi)$ (see Fig. 1) is

$$dn = \frac{n}{4\pi} \sin \theta d\theta d\varphi.$$

The total free energy W for the entire network is then obtained by integration of w according to (3) over the unit sphere, i.e.

$$W = \int w dn,$$

while the network principal stresses are formally obtained from (2), thus leading to

$$\sigma_i = \frac{C^R \sqrt{N} \pi^{2\pi}}{4\pi} \int_0^{\pi} \int_0^{2\pi} \mathcal{L}^{-1} \left(\frac{\lambda}{\sqrt{N}} \right) \frac{\lambda_i^2 m_i^2}{\lambda} \sin \theta d\theta d\varphi - p \quad (\text{no sum}) \quad (4)$$

where $C^R = nkT$ is known as the rubbery modulus. Once these principal stresses are evaluated, the stress tensor $\boldsymbol{\sigma}$ itself is constructed by application of (1).

It is noted that the above derivation is similar to that given by Treloar and Riding [3], but they limited their attention to *i)* biaxial deformations, and *ii)* to deformations with fixed principal axes of stretching so that the Eulerian and Lagrangian triads retained fixed orientations in space. The present formulation is valid for arbitrary three-dimensional deformations; for the biaxial deformations mentioned, the model reduces to that in [3].

Simplified Network Models

Now consider two simplified network models that have been proposed in the literature, namely the three-chain model and the eight-chain model. The three-chain model was originally suggested in [1] and assumes that a network containing of n chains per unit volume is equivalent to three independent sets of $n/3$ chains per unit volume parallel to the Eulerian principal axes as shown in Fig. 2a. The principal values of the stress tensor according to this model are

$$\sigma_i^{3\text{-ch}} = \frac{C^R \sqrt{N}}{3} \lambda_i \mathcal{L}^{-1}\left(\frac{\lambda_i}{\sqrt{N}}\right) - p \quad (\text{no sum}).$$

The eight-chain model was proposed in [6] and considers a set of eight chains connecting the central junction point and each of the eight corners of the unit cube as shown in Fig. 2b. The principal values of the stress tensor according to this model are given by

$$\sigma_i^{8\text{-ch}} = \frac{C^R \sqrt{N} \lambda_i^2}{3 \lambda} \mathcal{L}^{-1}\left(\frac{\lambda}{\sqrt{N}}\right) - p \quad (\text{no sum})$$

with

$$\lambda^2 = \frac{1}{3} \sum_{i=1}^3 \lambda_i^2.$$

As will be discussed briefly in the next section, the integration involved in (4) is a rather time-con-

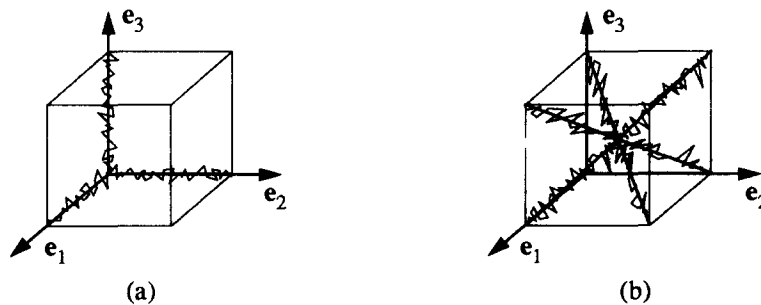


FIG. 2

Schematics of the three-chain model (a) and the eight-chain model (b).

suming procedure, which is certainly not appealing when one wishes to incorporate the model in for instance finite element computations. Furthermore, we shall demonstrate that the stress response predicted by (4) is always in between that predicted by the 3-chain model and the 8-chain model respectively. We propose to exploit this property to obtain an approximation of the full integration by combining the 3-chain and 8-chain models. One possibility is a simple linear combination,

$$\sigma_i = (1 - \rho) \sigma_i^{3\text{-ch}} + \rho \sigma_i^{8\text{-ch}}, \quad (5)$$

where the parameter ρ may be a constant or related to some other physical quantity which is for instance related to the deformation process. In this paper, we consider ρ to be related to the maximal principal stretch $\lambda_{\max} = \max(\lambda_1, \lambda_2, \lambda_3)$ by $\rho = 0.85 \lambda_{\max} / \sqrt{N}$ in which the factor 0.85 was chosen to give the best correlation with full integration of (4). In this way, the 8-chain contribution in (5) becomes increasingly important when λ_{\max} approaches the limit stretch \sqrt{N} .

Results

In Figs. 3 to 5 we present the stress responses (normalized by the rubber modulus C^R) for three well-known deformation processes, namely uniaxial tension, uniaxial compression and plane strain simple shear, for a material with $N = 36$. The hydrostatic pressure p is determined in the first two problems from the boundary condition that the lateral stresses vanish. At each stage of the deformation, the double integral in the full network relation (4) is evaluated by first noting that due to symmetry only the intervals $\theta \in [0, \pi/2]$, $\varphi \in [0, \pi/2]$ need to be considered, then subdividing that area into a number of cells and integrating within each cell using Gaussian quadrature. Recursive refinement is applied for each cell until the integral is obtained with a relative error of 10^{-5} .

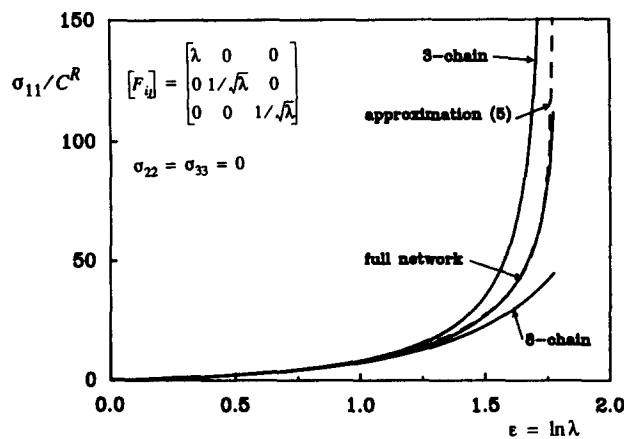


FIG. 3

Stress response to uniaxial tension for $N = 36$.

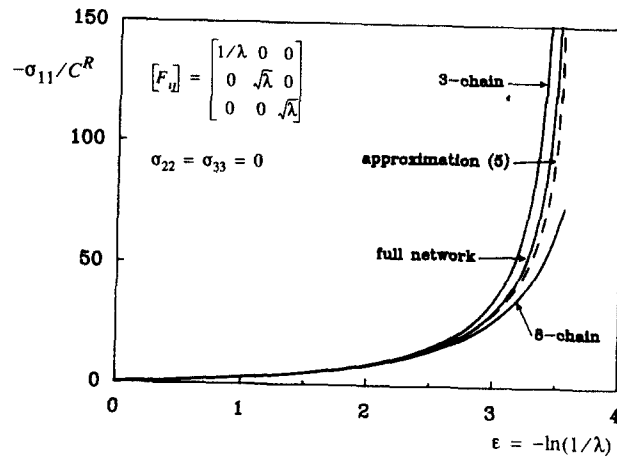


FIG. 4

Stress response to uniaxial compression for $N = 36$.

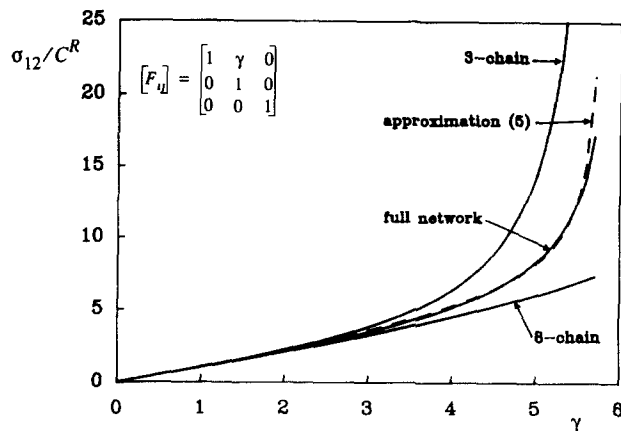


FIG. 5

Shear stress response to plane strain simple shear for $N = 36$.

As anticipated, the 8-chain model proposed by Arruda and Boyce [6] underestimates the actual stiffness of the network, whereas the more traditional 3-chain model provides an upper bound. The difference between the 3- and 8-chain models remains rather small up to roughly 50% of the limit stretch, but increases strongly when the limit stretch is approached. The proposed interpolation (5) captures the full network response very accurately up to very large stretches.

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